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DIFFUSION LIMIT FOR THE TRANSPORT OF QUASIFREE ELECTRONS IN LIQUIDS

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Abstract

A model of photoelectron emission by liquids is treated in terms of a transport problem. Its asymptotic behavior is studied in the limit when the mean free path becomes small and the geminetric recombination is neglected. This solution is then shown to be in agreement with that previously assumed in the Nemec theory.

1. INTRODUCTION

The model of photoelectron emission by liquids studied[1,2] involves three steps: generation of quasifree electrons in the bulk of the liquid, their transport towards the liquid-vacuum interface, and the overcoming of the barrier at the liquid-vacuum interface. The probability distribution of finding a quasifree electron generated at a certain point $\vec{r}_o$ with kinetic energy E_g, in a volume element $d\vec{r}$ about $\vec{r}$ with kinetic energy dE about E, plays a central role in the study of the energy distribution curves (EDC) of photoelectrons emitted by solutions. Such probability distributions can be obtained from the solution of a steady linear transport equation. We study

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the asymptotic behavior of such solution in the limit when the mean free path between collisions of the electron with the liquid molecules is small (diffusion theory limit) and the Coulombic interaction between the electron and its parent is not considered. Here we make use of known results from the neutron theory of slowing down problems (Fermi-age theory).[3] The solution we then obtain agrees in form with the solution assumed by Nemec.[2] In our case the law of energy loss in the process can be derived once we know the form of the electronic collision cross-section. The problem treated here, but with the added complication of geminate recombination, is presently being investigated by adapting different techniques.[4-6]

2. FORMULATION

We assume:

(a1) the electrons are classical point particles of mass m, moving about a medium consisting of host particles of mass M, with $m << M$;

(a2) the density of the electrons is sufficiently small so that the effect and frequency of electron-electron collisions are negligible;

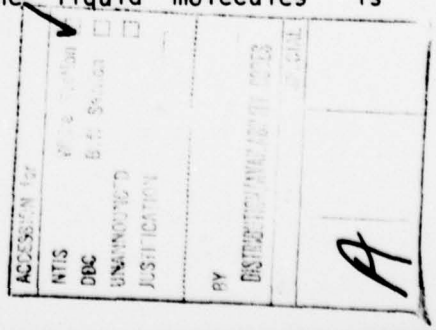
(a3) the solute molecules are sufficiently dilute in such a way that electron-ion interactions are negligible;

(a4) the liquid is isotropic and homogeneous;

(a5) there is no absorption (of electrons);

(a6) the coulombic interaction between the electron and its parent is not considered;

(a7) the scattering between electrons and the liquid molecules is isotropic.



Under the above assumptions (a1)-(a7) our photoelectron emission model can be formulated in terms of a steady linear transport equation for the electrons

$$[\vec{v}\cdot\vec{\nabla}_r + v\sigma(v)]N(r,v) = \int v'\sigma(v')f(v'\to v)N(\vec{r},\vec{v}')d\vec{v}' + S(\vec{r},\vec{v}) \qquad (2.1)$$

where $N(\vec{r},\vec{v})d\vec{r}d\vec{v}$ denotes the number of electrons in the element of volume $d\vec{r}d\vec{v}$ about $(\vec{r},\vec{v})$; $v\sigma(v)$ denotes the probability per unit time of collision of an electron whose speed is v; $f(v'\to v)dv$ the probability that an electron with speed v' before collision achieves a speed in the element dv about v; and S is a source term. Note because of (a5) $\int_0^\infty f(v'\to v)dv = 1$.

We are interested in the solution of (2.1) when an isotropic source is positioned at a point $\vec{r}_0$, which we can take to be the origin. Here we assume electrons disappear once their kinetic energy is below a certain specified value. This value can be its thermal energy or any other related to the interface barrier (liquid-vacuum) which is assumed to be constant.

Let $\vec{v} = v\vec{\Omega}$. The transport equation (2.1) can be rewritten as an integral equation for the electron density

$$n(\vec{r},v) = \int N(\vec{r},v\vec{\Omega})d\vec{\Omega} \qquad (2.2)$$

To do this, first integrate (2.1) along its characteristic curve in the following way. Denote the right-hand side of (2.1) by $Q(\vec{r},v)$, since $\vec{\Omega}\cdot\vec{\nabla}_r$ is simply the derivative taken along the direction of $\vec{\Omega}$, if we replace $\vec{r}$ by $\vec{r} - R\vec{\Omega}$ in (2.1) we can write it in the following form

$$[-d/dR + \sigma(v)]N(\vec{r}-R\vec{\Omega},v\vec{\Omega}) = v^{-1}Q(\vec{r}-R\vec{\Omega},v) \qquad (2.3)$$

after dividing both sides by v. If $Q(\vec{r}-R\vec{\Omega},v)$ is supposed known, integration of (2.3) leads to

$$N(\vec{r},v\vec{\Omega}) = N(\vec{r}-R_0\vec{\Omega},v\vec{\Omega})\exp\{-\sigma(v)R_0\} + v^{-1}\int_0^{R_0} Q(\vec{r}-R\vec{\Omega},v)\exp\{-\sigma(v)R'\}dR' \qquad (2.4)$$

after setting R = 0. Because there are no electrons coming directly from infinity we have

$$\lim_{R_0 \to \infty} N(\vec{r}-R_0\vec{\Omega},v\vec{\Omega})\exp\{-\sigma(v)R_0\} = 0.$$

Therefore we can write (2.4) as

$$N(\vec{r},v\vec{\Omega}) = v^{-1}\int_0^\infty Q(\vec{r}-R\vec{\Omega},v)\exp\{-\sigma(v)R\}dR \qquad (2.5)$$

If we integrate (2.5) over all Ω, put $\vec{r}-R\vec{\Omega} = \vec{r}'$ and notice that $dRd\vec{\Omega} = d\vec{R}/R^2 = d\vec{r}'/|\vec{r}-\vec{r}'|^2$, we find for (2.2)

$$n(\vec{r},v) = v^{-1}\int d\vec{r}'|\vec{r}-\vec{r}'|^{-2}Q(\vec{r}',v)\exp\{-\sigma(v)|\vec{r}-\vec{r}'|\}$$

or in terms of the flux $\phi(\vec{r},v) = vn(\vec{r},v)$

$$\phi(\vec{r},v) = (4\pi)^{-1}\int d\vec{r}'|\vec{r}-\vec{r}'|^{-2}\exp\{\sigma(v)|\vec{r}-\vec{r}'|\}\cdot$$
$$\{\int_0^\infty dv'\sigma(v')f(v'\to v)\phi(\vec{r}',v') + 4\pi S(\vec{r}',v)\} \qquad (2.6)$$

Equation (2.6) can be written in terms of energy dependent qualities by changing variables, $E = mv^2/2$. We get

$$\phi(\vec{r},E) = (4\pi)^{-1}\int d\vec{r}'|\vec{r}-\vec{r}'|^{-2}\exp\{-\sigma(E)|\vec{r}-\vec{r}'|\}\cdot$$
$$\{\int_0^\infty dE'\sigma(E')f(E'\to E)\,(\vec{r}',E') + 4\pi S(\vec{r}',E)\} \qquad (2.7)$$

where $f(v'\to v) = (2mE')^{1/2}f(E'\to E)$.

3. DIFFUSION WITH MODERATION APPROXIMATION

Assume the energy after each collision is, with equal probability, anywhere between $(1-\alpha)E'$ and E', for some fixed value of $\alpha > 0$, i.e.,

$$f(E'\to E) = \begin{cases} 1/\alpha E' & \text{for } (1-\alpha)E' \leq E \leq E' \\ 0 & \text{otherwise} \end{cases}$$

Then we can write (2.7) as

$$\phi(\vec{r},E) = (4\pi)^{-1}\int d\vec{r}'|\vec{r}-\vec{r}'|^{-2}\exp\{-\sigma(E)|\vec{r}-\vec{r}'|\}\cdot$$
$$\{\int_E^{E/(1-\alpha)} dE'(\alpha E')^{-1}\sigma(E')\phi(\vec{r}',E') + 4\pi S(\vec{r}',E)\} \qquad (3.1)$$

In terms of the lethargy $u = \ln(E_o/E)$, where E_o is so chosen that the lethargy would be positive for practically all the electrons which play a role in the theory, for example, $E_o = 100$ eV, and the collision density $F(\vec{r},u) = E\sigma(E)\phi(\vec{r},E)$, eq. (3.1) becomes

$$F(\vec{r},u) = (4\pi)^{-1}\sigma(u)\int d\vec{r}' \,|\vec{r}-\vec{r}'|^{-2}\exp\{-\sigma(u)|\vec{r}-\vec{r}'|\}\cdot$$

$$\{\alpha^{-1}\int_{u-\zeta}^{u} du' e^{u'-u}F(\vec{r}',u') + 4\pi S_u(\vec{r}',u)\} \qquad (3.2)$$

where $\zeta = -\ln(1-\alpha)$ and $S_u(\vec{r},u) = ES(\vec{r},E)$ is the number of electrons liberated per unit volume and unit time and per unit lethargy.

Equation (3.2) can be transformed by Taylor's expansion, into a differential equation if we assume S and F do not vary much within a mean free path $1/\sigma$. Expand F in powers of $(u'-u)$, then

$$F(\vec{r}',u') = F(\vec{r}',u) - (u-u')(\partial/\partial u)F(\vec{r}',u) + \text{higher order terms} \qquad (3.3)$$

Because

$$\alpha^{-1}\int_{u-\zeta}^{u} du' e^{u'-u} = 1$$

and

$$\alpha^{-1}\int_{u-\zeta}^{u} du'(u-u')e^{u'-u} = \alpha^{-1}\{-\zeta e^{-\zeta} - e^{-\zeta} + 1\}$$

$$= 1 + \alpha^{-1}(1-\alpha)\ln(1-\alpha) = \xi$$

(ξ = average increase in u), from (3.3) we obtain

$$\alpha^{-1}\int_{u-\zeta}^{u} du' e^{u'-u}F(\vec{r}',u') = F(\vec{r}',u) - (\partial/\partial u)\xi F(\vec{r}'u) \qquad (3.4)$$

if we neglect the higher order terms. The variation of F with respect to u is caused by its variation with $\vec{r}$ and it will be small if the latter is also small. This is a condition for the validity of this development.

Expand $F(\vec{r}',u)$ in powers of $(\vec{r}-\vec{r}')$,

$$F(\vec{r}',u) = F(\vec{r},u) + \sum_{i=1}^{3}(\vec{r}-\vec{r}')_i(\partial/\partial r_i)F(\vec{r},u)$$

$$+ 2^{-1}\sum_{i=1}^{3}\sum_{j=1}^{3}(\vec{r}-\vec{r}')_i(\vec{r}-\vec{r}')_j(\partial^2/\partial r_i\partial r_j)F(\vec{r},u)$$

$$+ \text{higher order terms} \qquad (3.5)$$

where the subscript i stands for the i-th component. Because of the factor $\exp\{-\sigma(u)|\vec{r}-\vec{r}'|\}$ in (3.2), only the contribution for small values of $|\vec{r}-\vec{r}'|$, up to the order of a few mean free paths $1/\sigma(u)$, will be significant. Therefore we replace $F(\vec{r}',u)$ in the first term of (3.4) by the first three terms of the series (3.5), $S_u(\vec{r}',u)$ and $(\partial/\partial u)\xi F(\vec{r}',u)$ by their values at $\vec{r}' = \vec{r}$. Since

$$(4\pi)^{-1}\sigma\int d\vec{r}'\,|\vec{r}-\vec{r}'|^{-2}\exp\{-\sigma|\vec{r}-\vec{r}'|\} = 1,$$

$$(4\pi)^{-1}\sigma\int d\vec{r}'\,|\vec{r}-\vec{r}'|^{-2}(\vec{r}-\vec{r}')_i\exp\{-\sigma|\vec{r}-\vec{r}'|\} = 0 \qquad (i = 1,2,3)$$

and

$$(4\pi)^{-1}\sigma\int d\vec{r}'\,|\vec{r}-\vec{r}'|^{-2}(\vec{r}-\vec{r}')_i(\vec{r}-\vec{r}')_j\exp\{-\sigma|\vec{r}-\vec{r}'|\} = (2/3\sigma^2)\delta_{ij}$$

we get from (3.2)

$$(\partial/\partial u)\xi F(\vec{r},u) = (1/3\sigma^2(u))\,\Delta_r F(\vec{r},u) + S_u(\vec{r},u) \tag{3.6}$$

4. COMPARISON WITH NEMEC'S APPROXIMATION

Introduce a new variable τ, referred to as the age of the electron,

$$\tau = \int_{u_o}^{u} du/3\xi\sigma^2(u) = \int_{E}^{E_o} dE/3\xi E\sigma^2(E) \tag{4.1}$$

The diffusion equation (3.6) then becomes

$$(\partial/\partial\tau)(\xi F) = \Delta_r(\xi F) + S_\tau(\vec{r},\tau) \tag{4.2}$$

where $S_\tau(\vec{r},\tau) = 3\xi\sigma^2(u)S_u(\vec{r},u)$ is the number of electrons produced by the source per unit time and unit τ interval.

For a point source of monoenergetic electrons and unit strength, equation (4.2) gives the distribution

$$q = \xi F = [4\pi(\tau-\tau_g)]^{-3/2}\exp\{-r^2/4(\tau-\tau_g)\} \tag{4.3}$$

where τ_g is the age of the electrons emitted by the source. If the source is not monochromatic, the solution of (4.2) is obtained by integrating (4.3)

against the distribution function of the energies or ages of the generated electrons.

The solution (4.3) here obtained agrees with Nemec's[2] if we identify the quantity he calls $R = (\ell\lambda/6)^{1/2}$, where ℓ is the total length of the random walk and λ is its mean free path, with the square root of $\tau-\tau_g$, i.e., if

$$R^2 = \tau - \tau_g = \int_E^{E_g} dE/3\xi E\sigma^2(E) \qquad (4.4)$$

If we assume $\sigma(E) = \sigma_o$, where σ_o is a constant, then from (4.4) we get

$$R = (3\xi\sigma_o^2)^{-1/2}[\ln(E_g/E)]^{1/2}$$

If $\sigma(E) = \sigma_o E^{-n/2}$, $n \neq 0$, then

$$R = (3\xi\sigma_o^2)^{-1/2}(E_g^n - E^n)^{1/2}$$

The laws of energy loss considered by Nemec can be recovered from the above for n = 3/2, 2 and 1

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